

FORM PTO-1390
(REV. 11-2000)

U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE

TRANSMITTAL LETTER TO THE UNITED STATES
DESIGNATED/ELECTED OFFICE (DO/EO/US)
CONCERNING A FILING UNDER 35 U.S.C. 371

ATTORNEY'S DOCKET NUMBER

31223-81205 (F-748)

U.S. APPLICATION NO. (If known, see 37 CFR 1.5)

09/937425

INTERNATIONAL APPLICATION NO.	INTERNATIONAL FILING DATE	PRIORITY DATE CLAIMED
PCT/EP00/02878	March 28, 2000	March 30, 1999
TITLE OF INVENTION Polypropylene Fibres		
APPLICANT(S) FOR DO/EO/US Axel Demain		

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371.
2. This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. 371.
3. This is an express request to begin national examination procedures (35 U.S.C. 371(f)). The submission must include items (5), (6), (9) and (21) indicated below.
4. The US has been elected by the expiration of 19 months from the priority date (Article 31).
5. A copy of the International Application as filed (35 U.S.C. 371(c)(2))
 - a. is attached hereto (required only if not communicated by the International Bureau).
 - b. has been communicated by the International Bureau.
 - c. is not required, as the application was filed in the United States Receiving Office (RO/US).
6. An English language translation of the International Application as filed (35 U.S.C. 371(c)(2)).
 - a. is attached hereto.
 - b. has been previously submitted under 35 U.S.C. 154(d)(4).
7. Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3))
 - a. are attached hereto (required only if not communicated by the International Bureau).
 - b. have been communicated by the International Bureau.
 - c. have not been made; however, the time limit for making such amendments has NOT expired.
 - d. have not been made and will not be made.
8. An English language translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).
9. An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)).
10. An English language translation of the annexes of the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).

Items 11 to 20 below concern document(s) or information included:

11. An Information Disclosure Statement under 37 CFR 1.97 and 1.98.
12. An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
13. A **FIRST** preliminary amendment.
14. A **SECOND** or **SUBSEQUENT** preliminary amendment.
15. A substitute specification.
16. A change of power of attorney and/or address letter.
17. A computer-readable form of the sequence listing in accordance with PCT Rule 13ter.2 and 35 U.S.C. 1.821 - 1.825.
18. A second copy of the published international application under 35 U.S.C. 154(d)(4).
19. A second copy of the English language translation of the international application under 35 U.S.C. 154(d)(4).
20. Other items or information:
PCT International Search Report

31223-81205
(F-748)

TRANSMITTAL LETTER
TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US)
FOR ENTRY INTO THE U.S. NATIONAL PHASE (Chapter 2)

INTERNATIONAL APPLICATION NO.	INTERNATIONAL FILING DATE	PRIORITY DATE CLAIMED
PCT/EP00/02878	March 28, 2000	March 30, 1999
TITLE OF THE INVENTION		
Polypropylene Fibres		
APPLICANT(S) FOR DO/EO/US		
Axel Demain		
PRIORITY APPLICATION NO.		
EP 99106462.7 (filed March 30, 1999)		

Via Express Mail

Box PCT
 Assistant Commissioner for Patents
 Washington, DC 20231

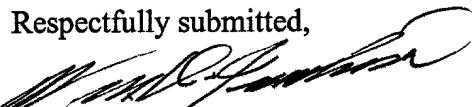
Dear Sir:

EXPRESS MAIL CERTIFICATE OF MAILING	
Express Mail Mailing Label No. EL 465 446 115 US	
I hereby certify that the papers enclosed herein are being deposited with the United States Postal Service "Express Mail Post Office to Addressee" service under 37 CFR 1.10 on the date indicated below and addressed to: BOX PCT, Assistant Commissioner for Patents, Washington, DC 20231.	
<i>Clare Jackson</i> Clare Jackson	September 26, 2001 Date of Deposit

Submitted herewith under cover of Form PTO-1390 is a copy of the International Application as filed (35 U.S.C. § 371(c)(2)). An English language translation of the application is not required since the International Application is being filed in English.

Submitted herewith is a check in the amount of \$1,260.00 as the fee for entry into the national phase as calculated on the attached Form PTC-1390. An oath or declaration of the inventor under 35 U.S.C. § 371(c)(4) will be submitted later.

Respectfully submitted,


 William D. Jackson
 Registration No. 20,846

Date: September 26, 2001

09/937425

JC16 Rec'd PCT/PTO SEP 26 2001

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POLYPROPYLENE FIBRES

F-748 PCT

The present invention relates to polypropylene fibres and to fabrics produced from polypropylene fibres.

Polypropylene is well known for the manufacture of fibres, particularly for manufacturing non woven fabrics.

EP-A-0789096 discloses such polypropylene fibres which are made of a blend of syndiotactic polypropylene (sPP) and isotactic polypropylene (iPP). That specification discloses that by blending from 0.3 to 3% by weight of sPP, based on the total polypropylene, to form a blend of iPP-sPP, the fibres have increased natural bulk and smoothness, and non-woven fabrics produced from the fibres have an improved softness. Moreover, that specification discloses that such a blend lowers the thermal bonding temperature of the fibres. Thermal bonding is employed to produce the non-woven fabrics from the polypropylene fibres.

The specification discloses that the isotactic polypropylene comprises a homopolymer formed by the polymerisation of propylene by Ziegler-Natta catalysis.

WO-A-96/23095 discloses a method for providing a non-woven fabric with a wide bonding window in which the non-woven fabric is formed from fibres of a thermoplastic polymer blend including from 0.5 to 25wt% of syndiotactic polypropylene. The syndiotactic polypropylene may be blended with a variety of different polymers, including isotactic polypropylene. The specification includes a number of examples in which various mixtures of syndiotactic polypropylene with isotactic polypropylene were produced. The isotactic polypropylene

comprised commercially available isotactic polypropylene, which is produced using a Ziegler-Natta catalyst. It is disclosed in the specification that the use of syndiotactic polypropylene widens the window of temperature over which thermal bonding can occur, and lowers the acceptable bonding temperature.

WO-A-96/23095 also discloses the production of fibres from blends including syndiotactic polypropylene which are either bi-component fibres or bi-constituent fibres. Bi-component fibres are fibres which have been produced from at least two polymers extruded from separate extruders and spun together to form one fibre. Bi-constituent fibres are produced from at least two polymers extruded from the same extruder as a blend. Both bi-component and bi-constituent fibres are disclosed as being used to improve the thermal bonding of Ziegler-Natta polypropylene in non-woven fabrics. In particular, a polymer with a lower melting point compared to the Ziegler-Natta isotactic polypropylene, for example polyethylene, random copolymers or terpolymers, is used as the outer part of the bi-component fibre or blended in the Ziegler-Natta polypropylene to form the bi-constituent fibre.

EP-A-0634505 discloses improved propylene polymer yarn and articles made therefrom in which for providing yarn capable of increased shrinkage syndiotactic polypropylene is blended with isotactic polypropylene with there being from 5 to 50 parts per weight of syndiotactic polypropylene. It is disclosed that the yarn has increased resiliency and shrinkage, particularly useful in pile fabric and carpeting. It is disclosed that the polypropylene blends display a lowering of the heat softening temperature and a broadening of the thermal response curve as measured by differential scanning calorimetry as a consequence of the presence of syndiotactic polypropylene.

US-A-5269807 discloses a suture fabricated from syndiotactic polypropylene exhibiting a greater flexibility than a comparable suture manufactured from isotactic polypropylene. The syndiotactic polypropylene may be blended with, *inter alia*, isotactic polypropylene.

EP-A-0451743 discloses a method for moulding syndiotactic polypropylene in which the syndiotactic polypropylene may be blended with a small amount of a polypropylene having a substantially isotactic structure. It is disclosed that fibres may be formed from the polypropylene. It is also disclosed that the isotactic polypropylene is manufactured by the use of a catalyst comprising titanium trichloride and an organoaluminium compound, or titanium trichloride or titanium tetrachloride supported on magnesium halide and an organoaluminium compound, *i.e.* a Ziegler-Natta catalyst.

EP-A-0414047 discloses polypropylene fibres formed of blends of syndiotactic and isotactic polypropylene. The blend includes at least 50 parts by weight of the syndiotactic polypropylene and at most 50 parts by weight of the isotactic polypropylene. It is disclosed that the extrudability of the fibres is improved and the fibre stretching conditions are broadened.

US-A-5648428 discloses a process for producing a polymer blend in a single reactor, specifically a polymer blend of isotactic polyolefin and syndiotactic polyolefin which may comprise polypropylene. The single reactor is provided with a catalyst system comprising a combination of at least one metallocene catalyst and at least one conventional supported Ziegler-Natta catalyst.

These known polypropylenes suffer from the disadvantage that the

processability of the iPP/sPP blends needs to be improved, particularly for spinning fibres, so that higher spinning speeds can be employed before fibre breakage occurs or the incidence of breakage at any given spinning speed is reduced.

It is further known to produce syndiotactic polypropylene using metallocene catalysts as has been disclosed for example in US-A-4794096.

WO-A-96/35729 discloses a process for preparing and using a supported metallocene-alumoxane catalyst which is used for the polymerisation or copolymerisation of olefins to produce an olefin polymer or copolymer having a broad and bimodal molecular weight distribution.

It is an aim of the present invention to produce a blend of iPP and sPP which has improved processability, particularly when forming spun fibres. It is a further aim of the present invention to provide fibres, and fabrics in particular non-woven fabrics made from those fibres, which have been produced from that blend.

The present invention provides a polypropylene blend including from 0.3 to 50% by weight of a syndiotactic polypropylene having a multimodal molecular weight distribution and at least 50% by weight of an isotactic polypropylene.

The sPP is typically bimodal.

The isotactic polypropylene (iPP) may have been produced using a Ziegler-Natta catalyst and may be a homopolymer or copolymer and may have a monomodal or multimodal molecular weight distribution.

Preferably, the syndiotactic polypropylene (sPP) concentration in the sPP/iPP blend is from 0.3 to 15wt%, more preferably from 1 to 10wt%. The fibre may be a two component sPP/iPP blend.

Preferably, the iPP is a homopolymer, copolymer, being either a random or block copolymer, or terpolymer of isotactic polypropylene.

Typically, the isotactic polypropylene has a melting temperature in the range of from 159 to 169°C, more typically from 161 to 165°C. The iPP employed in accordance with the invention has a molecular weight distribution typically having a dispersion index D of from 3.5 to 9, more preferably from 3.5 to 6.5. The dispersion index D is the ratio M_w/M_n , where M_w is the weight number average molecular weight and M_n is the number average molecular weight of the polymer. The iPP typically has a peak in the molecular weight distribution of around 35,000 to 60,000 kDa. The isotactic polypropylene may have an M_n of from 35,000 to 45,000 kDa.

The isotactic polypropylene preferably has a melt flow index (MFI) of from 1 to 90g/10mins, more preferably from 10 to 60g/10mins. In this specification the MFI values are those determined using the procedure of ISO 1133 using a load of 2.16kg at a temperature of 230°C.

The properties of two typical iPP resins for use in the invention are specified in Table 1.

The multimodal sPP, preferably bimodal sPP, is preferably a homopolymer or a random copolymer with a comonomer content of

from 0.1 to 1.5wt%, more preferably from 0.1 to 1wt%. The multimodal sPP may however be a block copolymer with a higher comonomer content or a terpolymer. If the comonomer content is above 1.5wt%, the sPP tends to become sticky, thus resulting in problems when spinning the fibres or thermally bonding the fibres. The comonomer content is selected so as to decrease the melting point of the sPP/iPP blend below 130°C. A lower melting point can also be obtained by using particular catalysts and/or process conditions during polymerisation of the sPP. Preferably, the sPP has a melting temperature of up to about 130°C. The sPP typically has two melting peaks, one being around 110°C and the other being around 125°C. The sPP typically has an MFI of from 0.1 to 1000g/10min, more typically from 1 to 60g/10min. The multimodal sPP may have an Mn of from 35,000 to 40,000 kDa. The properties of a typical bimodal sPP for use in the invention are specified in Table 1.

The sPP has a slightly narrower molecular weight distribution than for the iPP, wherein typically D may be from 3 to 6, more typically around 4 and has a peak of the molecular weight distribution at around 20,000 to 35,000 kDa. The sPP and the iPP may have substantially similar values for Mn. In view of the closeness between the peaks and the overlap of the molecular weight distributions of the sPP and the iPP, those two components can readily be blended together.

The present invention further provides a fibre produced from the polypropylene blend in accordance with the invention. The present invention further provides a fabric produced from the polypropylene fibre of the invention.

The present invention yet further provides a product including

that fabric, the product being selected from among others a filter, personal wipe, diaper, feminine hygiene product, incontinence product, wound dressing, bandage, surgical gown, surgical drape and protective cover.

The present invention is predicated on the discovery by the present inventor that when blended with iPP, multimodal sPP, preferably bimodal sPP, improves the processability of the polypropylene, particularly to form fibres in a spinning process, as compared to monomodal sPP.

The present inventor has found that when used in an amount of only about 2wt% sPP in the sPP/iPP blend, the spinnability is increased when sPP is multimodal, preferably bimodal.

The use of multimodal e.g. bimodal sPP in blends with iPP in accordance with the invention tends to provide fibres which can be more readily spun as compared to known fibres comprised of iPP blended with monomodal sPP. The molecular weight distributions of iPP and the multimodal sPP overlap, and as a result of the provision of sPP having a broad multimodal molecular weight distribution this provides increased overlap between the two distributions as compared to monomodal sPP, thereby improving the blend, and also provides a higher proportion of short chains in the combined molecular weight distribution for the blend. This has been found to result in an increase in the maximum spin speed before breakage for the fibres of the multimodal sPP/iPP blends in accordance with the invention.

The fibres produced in accordance with the invention may be either bi-component fibres or bi-constituent fibres. For bi-component fibres, iPP and sPP are fed into two different extruders. Thereafter the two extrudates are spun together to

form single fibres. For the bi-constituent fibres, blends of sPP/iPP are obtained by: dry blending pellets, flakes or fluff of the two polymers before feeding them into a common extruder; or using pellets or flakes of a blend of sPP and iPP which have been extruded together and then re-extruding the blend from a second extruder.

When the blends of multimodal sPP/iPP are used to produce fibres in accordance with the invention, for the production of spunlaid fibres, a typical extrusion temperature would be in the range of from 200°C to 260°C, most typically from 230°C to 250°C. For the production of staple fibres, a typical extrusion temperature would be in the range of from 230°C to 330°C, most typically from 280°C to 300°C.

The fibres produced in accordance with the invention may be produced from multimodal sPP/iPP blends having other additives to improve the mechanical processing or spinnability of the fibres. The fibres produced in accordance with the invention may be used to produce non-woven fabrics for use in filtration; in personal care products such as wipers, diapers, feminine hygiene products and incontinence products; in medical products such as wound dressings, surgical gowns, bandages and surgical drapes; in protective covers; in outdoor fabrics and in geotextiles. Non-woven fabrics made with the bimodal sPP/iPP fibres of the invention can be part of such products, or constitute entirely the products. As well as making non-woven fabrics, the fibres may also be employed to make a woven knitted fabric or mat. The non-woven fabrics produced from the fibres in accordance with the invention can be produced by several processes, such as air through blowing, melt blowing, spun bonding or bonded carded processes. The fibres of the invention may also be formed as a

non-woven spunlace product which is formed without thermal bonding by fibres being entangled together to form a fabric by the application of a high pressure-fluid such as air or water.

The present invention will now be described in greater detail with reference to the following non-limiting example.

Example 1 and Comparative Example 1

In accordance with Example 1, a bimodal sPP having a dispersion index D of 3.9 and an MFI of 4.4g/10min was blended in an amount of 2wt% based on the weight of the blend with an isotactic polypropylene having the properties of Polymer 1 specified in Table 1. The blend was then subjected to spinning through a spinnerette to form fibres and the maximum spinning speed before breakage of the fibres was measured at two spinning temperatures.

The results are shown in Table 2.

In accordance with Comparative Example 1, a monomodal sPP having a dispersion index D of 2.6 and an MFI of 4.5g/10min, i.e. an MFI substantially the same as that of the bimodal sPP of Example 1, was blended, also in an amount of 2wt% based on the weight of the blend, with the same isotactic polypropylene employed in Example 1. The resultant blend was similarly spun to form fibres at the same temperatures as for Example 1 and the results are also shown in Table 2.

It may be seen that for both spinning temperatures of 260°C and 280°C, the use of bimodal sPP substantially increases the maximum spinning speed before breakage of the fibres as compared to the use of the same amount of monomodal sPP blended with the same isotactic polypropylene.

TABLE 1

		Bimodal sPP	iPP - Polymer 1	iPP Polymer 2
MI ₂	g/10min	3.6	25.9	14
T _m	°C	110 and 127	163.4	164.3
M _n	kDa	37426	35756	41983
M _w	kDa	160229	192383	256895
M _z	kDa	460875	755427	1173716
M _p	kDa	50516	96230	107648
D		4.3	5.4	6.1

TABLE 2

	Example 1	Comparative Example 1
Property	iPP-2wt% bimodal sPP	iPP-2wt% monomodal sPP
Maximum spinning speed at 260°C	3700	3300
Maximum spinning speed at 280°C	3100	2900

CLAIMS:

1. A polypropylene blend including from 0.3 to 50% by weight of a syndiotactic polypropylene having a multimodal molecular weight distribution and at least 50% by weight of an isotactic polypropylene.
2. A blend according to claim 1 wherein the multimodal sPP concentration in the sPP/iPP blend is from 0.5 to 15wt%.
3. A blend according to claim 2 wherein the multimodal sPP concentration in the sPP/iPP blend is from 1 to 10wt%.
4. A blend according to any foregoing claim wherein the iPP is a homopolymer, copolymer or terpolymer of isotactic polypropylene.
5. A blend according to any foregoing claim wherein the iPP has a dispersion index (D) of from 3.5 to 9, preferably 3.5 to 6.5.
6. A blend according to any foregoing claim wherein the iPP has a melting temperature in the range of from 159 to 169°C.
7. A blend according to any foregoing claim 4 to 6 wherein the iPP has an Mn of from 35,000 to 60,000 kDa.
8. A blend according to any foregoing claim wherein the iPP has a melt flow index (MFI) of from 1 to 90g/10mins.
9. A blend according to any foregoing claim wherein the multimodal sPP is a homopolymer or a random or block copolymer or a terpolymer.

10. A blend according to any foregoing claim wherein the multimodal sPP has a melting temperature of up to about 130°C.

11. A blend according to any foregoing claim wherein the multimodal sPP has an MFI of from 0.1 to 1000g/10min.

12. A blend according to claim 11 wherein the multimodal sPP has an MFI of from 1 to 60g/10min.

13. A blend according to any foregoing claim wherein the multimodal sPP has a Mn of from 35,000 to 40,000 kDa.

14. A blend according to any foregoing claim wherein the multimodal sPP has a dispersion index (D) of from 3 to 6.

15. A blend according to any foregoing claim wherein the multimodal sPP is bimodal.

16. A spun polypropylene fibre produced from the blend of any foregoing claim.

17. A fabric produced from the polypropylene fibre according to claim 16.

18. A product including a fabric according to claim 17, the product being selected from a filter, personal wipe, diaper, feminine hygiene product, incontinence product, wound dressing, bandage, surgical gown, surgical drape and protective cover.

19. Use, for increasing the maximum spinning speed when producing spun polypropylene fibres, of from 0.5 to 50wt% multimodal syndiotactic polypropylene in a blend with isotactic polypropylene.



PATENT APPLICATION
Docket No.: F-748

DECLARATION

As below named inventor, I, AXEL DEMAIN, hereby declare that:

My residence address and citizenship are as stated below next to my name;

I believe I am the original, first and sole inventor of the subject matter which is described and claimed and for which a patent is sought on the invention, design or discovery entitled: POLYPROPYLENE FIBRES, the specification of which was filed on March 28, 2000, as International Application No. PCT/EPO 00/002878, and on Sept. 26, 2001, as U.S. Application Serial No. 09/937,425.

I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above;

I do not know and do not believe that said invention, design or discovery was ever known or used in the United States of America before our invention or discovery thereof, or patented or described in any printed publication in any country before our invention or discovery thereof, or more than one year prior to this application, or in public use or on sale in the United States of America more than one year prior to this application; that said invention, design or discovery has not been patented or made the subject of an inventor's certificate issued prior to the date of this application in any country foreign to the United States of America on an application filed by my legal representatives or assigns; and that I acknowledge my duty to disclose information of which I am aware which is material to patentability in accordance with 37 C.F.R. Section 1.56(a).

I hereby claim foreign priority benefits under 35 U.S.C. Section 119 of the foreign application(s) for patent or inventor's certificate listed below and have also identified below the foreign application for patent or inventor's certificate having a filing date before that of the application for which priority is claimed:

Number	Country	Date Filed
99106462.7	EPO	March 30, 1999

Please send correspondence to:
to:

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Please direct telephone calls

David J. Alexander
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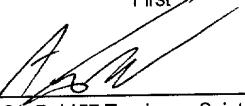
Serial No. 09/937,425
Attorney Docket No. F-748

Declaration - Axel Demain
Page 2 of 2

I hereby declare that all statements made herein of our own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

1-00) Full name of first inventor

Axel A G F Demain
First Middle Last

First inventor's signature 

Residence: Rue de Sart, 21, B-1457 Tourinnes-Saint-Lambert, Belgum

BEX.

Date 19 Nov 2001

Citizenship: Belgium



PATENT APPLICATION
F-748

POWER OF ATTORNEY

We, ATOFINA Research, a corporation of the country of Belgium and having its principal offices at Zone Industrielle C, 7181 Seneffe (Feluy), Belgium, ASSIGNEE of the entire interest in the invention, application and any Letters Patent that may issue thereon for POLYETHYLENE FIBRES, the specification of which was filed on Sept. 26, 2001, as Application Serial No. 09/937,425, hereby appoint:

8 -
David J. Alexander, Registration No. 33,221
WILLIAM D. JACKSON, Registration No. 20,846;
L. DAN TUCKER, Registration No. 22,670;
ROY W. HARDIN, Registration No. 28,304;
MARTIN KORN, Registration No. 28,317;
MONTY L. ROSS, Registration No. 28,899;
CHARLES PHIPPS, Registration No. 40,127; and
MICHAEL DUBNER, Registration No. 47,310.

as our attorney with full power of substitution and revocation, to prosecute this application and to transact all business in the United States Patent and Trademark Office connected therewith, and to file and prosecute any international or foreign patent applications filed thereon before any international or foreign authorities.

Please send correspondence and direct phone calls to:

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Attorney Docket No. F-748

We declare that the below-named individual is authorized to execute this Power of Attorney on behalf of ASSIGNEE.

PATENT APPLICATION
F-748

ASSIGNEE CERTIFICATION

In accordance with 37 CFR §3.73 the ASSIGNEE hereby certifies that the evidentiary documents with respect to its ownership have been reviewed and that, to the best of ASSIGNEE's knowledge and belief, title is in the ASSIGNEE seeking to take this action.

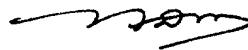
ATOFINA RESEARCH

Signature: _____

Name: _____

Title: _____

Date: 15 October 2001 16.10.2001



Patient Formalities
Officer.

J.C. DEWART
Department
Manager